



The influence of the network topology on the deformation and fracture behaviour of silica glass: A molecular dynamics study

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ABSTRACT

The nano-structural arrangement of silica glass can be analysed based on its short- and medium-range order. The latter is represented by rings of various sizes composed of corner-sharing tetrahedra. In this paper, a topologically motivated explanation of the mechanical behaviour of silica glass is provided. We perform classical molecular dynamics simulations to investigate the influence of the ring distribution on the deformation and fracture behaviour depending on the quenching rate. For this purpose, we prepare several samples with statistically different vitreous states by quenching molten silica to ambient temperature using various quenching rates. All samples are subjected to tensile loadings, based on which we demonstrate that the mechanical behaviour depends significantly on the associated ring statistics.

1. Introduction

Silica glass or vitreous silica ($v\text{-SiO}_2$) is one of the most used materials in many technological fields, such as electrical engineering, electronics, communication technology, optics, chemical engineering and metallurgical industries [1,2]. Many scientific disciplines make efforts to extend the understanding of $v\text{-SiO}_2$. In particular, the understanding of its mechanical properties and fracture mechanism is of high interest and subject of recent research [3–9]. It is known that cracks propagate rapidly through brittle materials as soon as a critical stress level is exceeded [10]. As the macroscopic observable fracture behaviour is driven by effects that occur on the nano-scale, atomistic studies are essential in order to get a deep insight into the physical nano-mechanisms and, hence, to interpret the macroscopic behaviour of the material.

Vitreous materials do not show a long-range structural order on the nano-scale. However, in the case of the SiO_2 compound, a description of the structural arrangement by means of its short- and medium-range order is possible [11]. In that case, silicon (Si) is fourfold coordinated and, thus, surrounded by four oxygen (O) atoms. On the one hand, these nearest-neighbour Si and O atoms form an elementary tetrahedron unit. On the other hand, two neighbouring tetrahedra share an O atom (corner-sharing, cf. Fig. 1), and these inter-connected tetrahedra assemble closed paths, which are known as n -membered rings of various sizes containing n silicon atoms [12,13].

The above-described network topology has been used within the framework of topological constraint theory in order to describe the

structural dependence of glass properties [14–19]. In this framework, the network of glasses is described in terms of nodes (atoms) connected through simplified mechanical trusses (chemical bonds) representing mechanical constraints. Based on this, a three-dimensional network is classified as flexible or under-constrained, if the number of constraints per atom N_c is lower than three (degrees of freedom), stress-rigid or over-constrained, if N_c is greater than three. Following this, networks that consist of both flexible and stress-rigid regions are called isostatic [20–22]. In this regard, extensive investigations have been performed on numerous glasses [20,23–26]. Using the constraint theory, the network topology has been shown to strongly affect macroscopic glass properties, such as hardness [27–29] and fracture behaviour [30,31].

Over the last decades, classical molecular dynamics (MD) simulations based on numerical integration of the Newtonian equations of motion for a set of interacting atoms have shown their efficacy in modelling materials on the nano-scale, especially in studying material properties, which are not easily accessible from experiments [32]. Many studies have been carried out to investigate the structural properties and the atomistic mechanical behaviour of silica glass. Usually, the preparation of $v\text{-SiO}_2$ is realised by quenching molten silica to ambient temperature [33–36]. Numerous simulations have been performed on multi-million atom models to research the origin of crack nucleation and propagation in both nano-structured and vitreous SiO_2 [34,37–41]. Other studies comprehensively investigated the deformation and fracture behaviour of $v\text{-SiO}_2$ under different simulation conditions, e.g. by varying the box dimensions, quenching rate or the strain rate during the loading process [36,42–44]. Furthermore, initial studies

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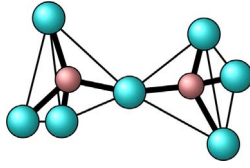


Fig. 1. Sketch of two corner-sharing tetrahedra, where the cyan balls represent oxygen and the red balls represent silicon atoms.

demonstrated significant effects of the quenching rate on the relative arrangement of tetrahedra [45]. Recent MD simulations have also shown a clear influence of the thermal treatment on the structural and mechanical properties for other vitreous materials, such as bioactive and metallic glasses [46–48]. Investigations that associate the n -membered rings with the mechanical behaviour of silica, e.g. to analyse the anisotropic deformation response or structural transformation induced by high pressure, can be found in [49–53].

Despite the large number of the above-mentioned studies on the structural properties and the mechanics of v -SiO₂, the influence of the medium-range order on the deformation and fracture behaviour caused by the quenching rate has, to date, not been investigated. Using a state-of-the-art ring search strategy realised within an in-house implementation, we demonstrate firstly that different quenching rates generate samples of v -SiO₂ with different medium-range orders. Subsequently, we show that the distribution of rings governs the mechanical behaviour on the nano-scale and, therefore, results in stress–strain relations which vary to a significant degree. Based on this, the focus is directed towards a connection between the distribution of rings and the mechanical behaviour under mode I tensile loading.

2. Simulation methods

The MD simulations of the preparation and deformation are performed with the open-source code LAMMPS (Large-scale Atomic/Molecular Massively Parallel Simulator) [54]. Numerous potential functions are available in the literature for the description of vitreous silica, including both purely two-body, and combined two- and three-body interactions [33,55–57]. It should be noted that the quality of the simulation results significantly depends on the choice of the potential function. In this paper, the inter-atomic forces are calculated due to a potential function consisting of two- and three-body covalent interactions [33]. This potential has extensively been used in numerical studies, especially in investigating the fracture mechanisms of vitreous silica, and validated, comparing simulation and experimental results for both structural and mechanical properties [12,33,34,37–41,50,58,59]. The two-body part describes the effects of steric repulsion, Coulombic charge transfer and electronic polarisabilities. The three-body part includes angular bond bending and bond stretching between silicon and oxygen atoms. The functional form of the potential $U = U_{ij} + U_{jik}$ is as follows:

$$U_{ij} = \frac{H_{ij}}{r_{ij}^{\eta_{ij}}} + \frac{Z_i Z_j}{r_{ij}} + \frac{1}{2} \frac{(\alpha_i Z_j^2 + \alpha_j Z_i^2)}{r_{ij}^4} \exp\left(\frac{-r_{ij}}{\lambda_{4,ij}}\right),$$

$$U_{jik} = B_{jik} f_{jik} (\cos \vartheta_{jik} - \cos \bar{\vartheta}_{jik})^2, \quad (1)$$

$$f_{jik} = \begin{cases} \exp\left(\frac{1}{r_{ij} - r_0} + \frac{1}{r_{ik} - r_0}\right) & \text{for } r_{ij}, r_{ik} < r_0, \\ 0 & \text{for } r_{ij}, r_{ik} > r_0. \end{cases}$$

Here, r_{ij} is the distance between atoms i and j , and r_{ik} is the distance between atoms i and k . The magnitudes H_{ij} and η_{ij} denote the repulsive strength and exponent, the magnitudes α_i and Z_i are the electronic polarisability and effective charge, respectively. The three-body part of the potential is defined within a cut-off distance r_0 . The terms B_{jik} and ϑ_{jik} represent the strength of the three-body part and the angle between the vectors $\mathbf{r}_{ij} = \mathbf{r}_i - \mathbf{r}_j$ and $\mathbf{r}_{ik} = \mathbf{r}_i - \mathbf{r}_k$, where $\mathbf{r}_i, \mathbf{r}_j$ and \mathbf{r}_k are the position

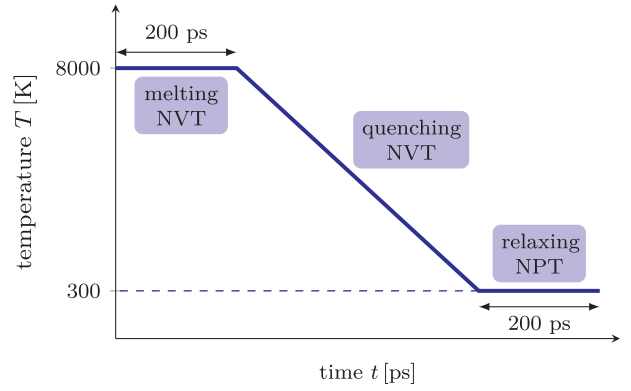


Fig. 2. Illustration of the melting-quenching process for the preparation of silica glass.

vectors of the atoms i, j and k , respectively. The constant $\bar{\vartheta}_{jik}$ is the equilibrium bond angle. For further explanation concerning the parameters of the respective pairs and triplets of atoms, we refer to [33,58].

During the MD simulations, the two-body interaction is calculated between neighbouring atoms that are within a cut-off distance of $r_c = 5.5 \text{ \AA}$, and the function is tilted and shifted so that both the potential energy and force vanish at r_c , as proposed in [60]. Furthermore, the simulations are carried out under periodic boundary conditions on cuboid systems with edge lengths of $l_x = 89 \text{ \AA}$, $l_y = 71 \text{ \AA}$, $l_z = 57 \text{ \AA}$, containing 24,000 atoms. The initial dimensions of the simulation box are chosen so that the initial mass density ρ_0 is 2.2 g/cm^3 [12,33].

Vitreous silica is prepared by a melting-quenching process [33–36]. Starting from a random spatial arrangement of the atoms, the system is melted at a temperature of 8000 K for 200 ps to reduce the influence of the initial conditions. Subsequently, the melted silica is quenched linearly to ambient temperature of 300 K, while the quenching is carried out with different cooling rates Q_i (cf. Fig. 2).

In this paper, five different quenching rates Q_i are chosen: $Q_1 = 1 \text{ K/ps}$, $Q_2 = 5 \text{ K/ps}$, $Q_3 = 25 \text{ K/ps}$, $Q_4 = 50 \text{ K/ps}$ and $Q_5 = 100 \text{ K/ps}$. It must be noted that the quenching rates used in this paper, are many orders of magnitude larger than those performable in the laboratory, as the quenching process is realised by heat exchange through the surfaces. Contrariwise, the dimensions of the simulation boxes are much smaller, and quenching is realised by a chain of thermostats, subjected to the sample volume. This considerably relativises the difference in time scale between simulations and experiments, as also pointed out in [28]. Nevertheless, the quenching rates are chosen based on previous studies [28,33,35,36,42–44,61,62], and qualitative comparisons between our simulation results and experimental studies are performed throughout this paper. Both the melting and quenching processes are performed under isothermal and isochoric (NVT) conditions. Finally, the silica glass is equilibrated at 300 K for additional 200 ps under isothermal and isobaric (NPT) conditions. The thermostating and barostating is realised by using a chain of three thermostats and three barostats based on the Nosé-Hoover equations. The Verlet algorithm is applied for the time integration. The silica glass specimens, obtained through the above-described procedure, are subsequently subjected to uniaxial deformation. The deformation is carried out in x -direction at a constant strain rate of $\dot{\epsilon}_{xx} = 0.001 \text{ ps}^{-1}$ applying NPT conditions at the temperature of 300 K and the pressure of 1 bar. The stress tensor σ is calculated via the Virial theorem by using the equation

$$\sigma = -\frac{1}{V} \sum_{i=1}^N [m_i \mathbf{v}_i \otimes \mathbf{v}_i + \mathbf{r}_i \otimes \mathbf{f}_i], \quad (2)$$

where V denotes the simulation box volume, N is the total number of atoms, m_i is the mass of the i th atom. The vectors \mathbf{r}_i and \mathbf{v}_i are the position and velocity vector, respectively, and \mathbf{f}_i is the force vector acting on the i th atom [54,63]. The symbol \otimes represents the dyadic product of two vectors.

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